CRYSTAL-LIQUID-VAPOR EQUILIBRIUM EXPERIMENTS AT HIGH TEMPERATURE ($\leq 1800^{\circ}$C) AND LOW, CONTROLLED OXYGEN AND HYDROGEN PRESSURE (10^{-1} - 10^{-9} PA)

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Evidence from carbonaceous chondrites points to refractory oxides in the system $CaO-MgO-Al_2O_3-TiO_2-SiO_2-Fe-O$ as being among the earliest phases to condense from the solar nebula (see MacKinnon and Rietmeijer, Condensation Kinetics. . ., This Report). Conversely, in condensation-sublimation models of the chemical evolution of the early solar nebula, the refractory phases will be the last to evaporate. Calculations of the relative mineral stabilities rely on untested assumptions regarding speciation in the gas phase and, sometimes, on less than accurate thermodynamic data for relevant condensed phases (e.g., liquid as well as solid calcium aluminates). It is, therefore, necessary to establish the equilibrium relationships between the relevant crystalline and amorphous phases before the chemical constraints can be meaningfully applied to models of solar system history.

Preliminary experiments on Earth show that such experiments are feasible. These experiments have been conducted on Earth with synthetic diopside and akermanite composition as starting materials. The results indicate that the p_{H2}-temperature conditions governing crystal-liquid, liquid-gas and crystal-gas equilibria might differ substantially from those suggested from calculated models. Lower pressures than attainable in earth-based furnace systems appear necessary. Moreover, incongruent vaporization of both liquids and crystals (leaving a calcium-enriched residue) has been observed.

Condensation experiments from a refractory-oxide containing gas phase in a controlled temperature gradient are highly desirable and can be conducted simultaneously with the phase equilibrium experiments with only minor modification of the presently operating furnace configuration. Finally, a mechanism to analyze the gas phase will provide add information not only to the phase equilibrium experiments but would also provide valuable thermodynamic data about the phases present.

Earth-based experiments suffer from several unavoidable problems. In order to attain controlled, relevant f_{O2} and f_{H2} (with $p_{H2} + p_{O2} = p_{total}$) very high vacuum, not attainable in terrestrial laboratories , is required. Preliminary tests in our terrestrial laboratory with melilite and pyroxene compositions indicate that in order to conduct adequate experiments on these phases as well as on calcium-magnesium aluminates, the ambient pressure should be less than 10^{-11}Pa. This pressure can be reached with the molecular shield technology. Furthermore, even for the best terrestrial vacuum conditions ($\geq 10^{-8}$ Pa), continuous pumping on the system is necessary, thus, rapidly removing gas components. Equilibrium is, therefore, difficult to ascertain. These problems can be overcome by experimentation in the Space Station where the experiments can be conducted under near static pressure conditions and where total pressure equals the sum of controlled hydrogen and oxygen pressures and be controlled for periods exceeding several hours (up to days).

The condensation and fractionation in the early solar nebula occurred as substantially lower gravity than attainable in high-temperature, low-pressure experiments on earth. Microgravity is available in the Space Station, thus, providing a more realistic environment in which to establish the equilibrium relationships.

Finally, terrestrial experiments suffer from container

problems where the container may interact chemically with the experimental charge and where the container material can impose an external, unwanted oxygen fugacity. Levitation techniques can provide container-free sample environments, thus, completely eliminating these problems.

Technical Requirements

The experiments require temperatures as high as 1800° C, to be controlled within $\pm 10^\circ$C to 1800°C. With the laboratory model, a 5 cm long, 1 cm diameter furnace is used for this purpose. The samples would be 5-10 mg. Power requirements are less than 100 Watts at 110V AC. Because of the low power and high vacuum conditions, thermal insulation is not a problem given the necessary experimental durations (several hours). The furnace is easily adapted to lower-voltage dc power.

Temperature control is accomplished with Tungsten-Rhenium thermocouples interfaced with the temperature control circuit. Hydrogen and oxygen gas pressures are maintained with conventional gas mixing methods. Both the gas pressure , sample change and pressure control can be easily modified for automatic and remote control. Sample-retrieval from, and loading of new, sample carousel every 7-14 days are appropriate. The pressures attainable with molecular shield technology are adequate.

On-site optical and scanning electron microscopic facility (EDS-equipped) are important as are real-time video and data transfer during sample examination. Mass-spectrometer for gas analysis and capability for optical and vibrational spectroscopy is also desirable.

The total volume and weight of the prototype, laboratory-tested model is about 4 kg and 4000 cm^3 (not including the vacuum system). These weight and volume requirements might

be reduced further depending on temperature and pressure automation techniques and whether or not an automated sample stage for remote sample insertion and retrieval is implemented.